

# Sub-MHz Linewidth at 240 GHz from an Injection-Locked Free-Electron Laser

Susumu Takahashi,\* Gerald Ramian, and Mark S. Sherwin  
*Department of Physics and Institute for Quantum and Complex Dynamics,  
 University of California, Santa Barbara CA 93106*

Louis-Claude Brunel and Johan van Tol  
*National High Magnetic Field Laboratory, Florida State University, Tallahassee FL 32310*  
 (Dated: February 1, 2008)

Radiation from an ultra-stable 240 GHz solid-state source has been injected, through an isolator, into the cavity of the University of California Santa Barbara (UCSB) MM-wave free-electron laser (FEL). High-power FEL emission, normally distributed among many of the cavity's longitudinal modes, is concentrated into the single mode to which the solid state source has been tuned. The linewidth of the FEL emission is 0.5 MHz, consistent with the Fourier transform limit for the 2 microsecond pulses. This demonstration of frequency-stable, ultra-narrow-band FEL emission is a critical milestone on the road to FEL-based pulsed electron paramagnetic resonance spectroscopy.

Scientific and technological opportunities in the electromagnetic spectrum between 0.1 and 10 terahertz (THz) have fueled rapid growth in the technology for generating THz radiation.<sup>1,2</sup> THz sources can now generate coherent broad-band radiation suitable for spectroscopic<sup>2</sup> and high-field nonlinear<sup>3,4</sup> experiments, as well as narrow-linewidth radiation at fixed<sup>2,5</sup> or tunable<sup>6</sup> frequencies. Free-electron lasers (FELs) are proven sources of tunable high-power THz radiation.<sup>7,8</sup> Almost all FELs are driven by radio-frequency linear accelerators (RF-LINACs), and emit pulses with a typical duration measured in picoseconds. Such pulses have relatively broad, Fourier-transform limited linewidths. The University of California Santa Barbara (UCSB) FELs are driven by an electrostatic accelerator. Under free-running operation, these FELs generate pulses which are several microseconds long but lase simultaneously on a number of modes.

An important niche has remained unfilled: tunable, high-power ( $> 100$  W) THz pulses with intrinsically narrow linewidths. Such sources are desirable for nonlinear experiments involving very narrow excitation lines, such as pulsed electron paramagnetic resonance (EPR), as well as nonlinear spectroscopy of dilute molecular gases and Rydberg atoms.

This letter shows that the frequency of UCSB's MM-wave FEL (120-890 GHz) can be locked to the frequency of an ultra-stable solid-state oscillator. The experiment demonstrates that the important niche for tunable, high-power, THz sources with intrinsically narrow linewidths can be filled using electrostatic-accelerator driven FELs. It also represents a key milestone on the road to high-power pulsed EPR at frequencies of 240 GHz and above.

The spectrum of a single pulse of the free-running FEL is shown in Fig. 1(a). The spectrum shows many modes with the 25 MHz spacing which correspond to longitudinal modes of the 6 m long FEL cavity. The FEL spectrum and center frequency are slightly different on each pulse (pulse-to-pulse fluctuation of center frequency  $\sim 0.05\%$ ). Pulses with spectra like these are routinely used for many experiments. However, pulsed EPR requires a much narrower spectrum.

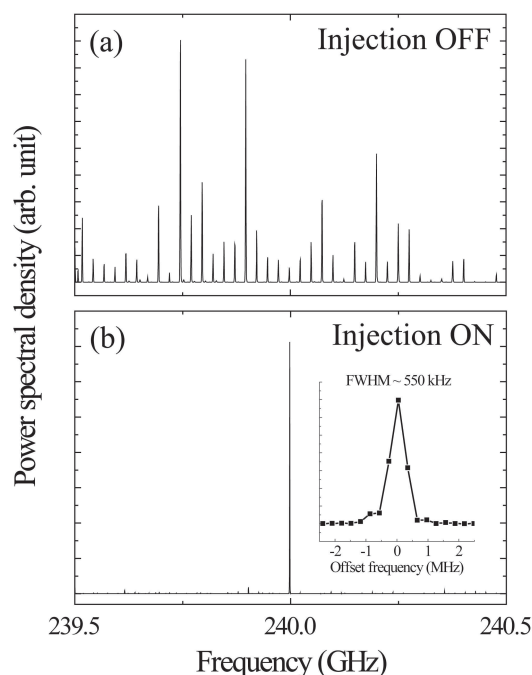


FIG. 1: FEL spectra. (a) FEL spectrum without the injection-locking. The spectrum shows many longitudinal modes spaced by 25 MHz. (b) FEL spectrum with injection source on. Single-mode lasing is observed with no pulse-to-pulse frequency fluctuations.

A well-known technique to force a laser to emit on a single mode is to inject into the laser cavity a weak seed beam from an oscillator with a very stable and well-defined frequency. This technique is called injection-locking. If the power of the seed beam is significantly larger than the power in the random electromagnetic fluctuations that are usually amplified for lasing, then only the seed beam will be amplified. Fig. 1(b) is the spectrum with injection on. The effect is immediate and unequivocal. Lasing always occurs at one frequency on the same

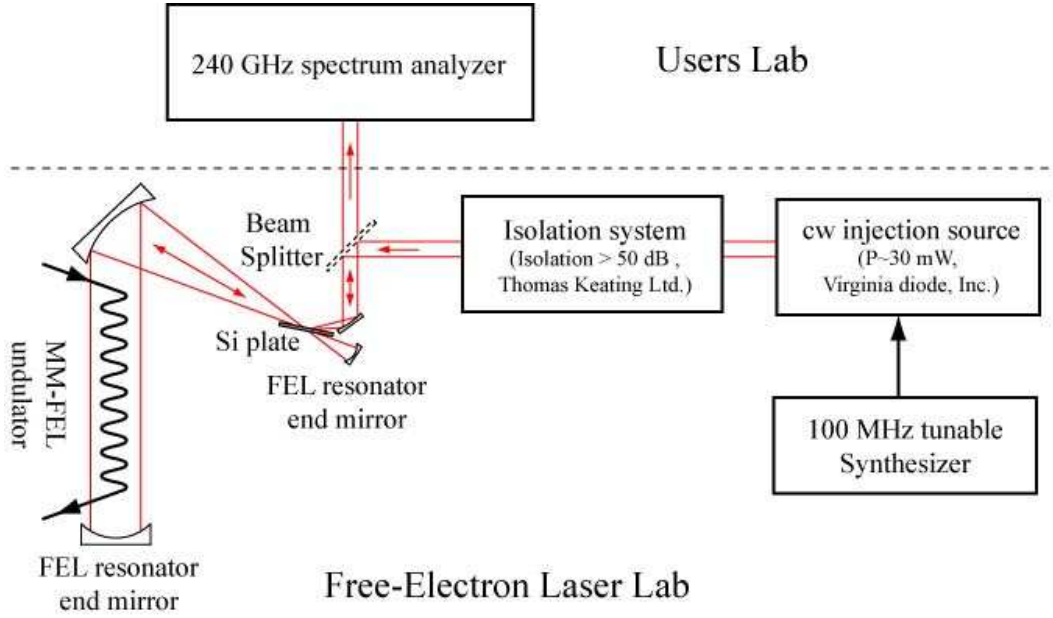


FIG. 2: Schematic of the injection-locking system for the UCSB MM-FEL. The injection source, isolator and tunable 100 MHz synthesizer are located in free-electron laser lab. The FEL outputs are sent to the users lab through a vacuum optical transport system.

longitudinal mode selected by the injection source frequency. The injection-locking is observed only when the source frequency matches one of the resonator's longitudinal modes. The full-width half maximum (FWHM) width of the lasing line is  $\sim 550$  kHz which is close to the Fourier transform limit of the sampling window ( $\sim 2\mu s$ ).

Fig. 2 illustrates the UCSB injection-locking system. The cw injection source, built by Virginia Diode Inc. (VDI),<sup>6</sup> generates as much as 30 mW at 240 GHz. The VDI source consists of a 15 GHz phase-locked oscillator, RF active frequency doubler, and three varactor doubler stages. The source frequency is tunable within small frequency range by employing a tunable 100 MHz digital synthesizer as a reference. This tunability is essential to match the injection source frequency to the FEL resonator and to facilitate operation of the injection-locking system. A key component is a quasi-optical Faraday-rotation, isolator developed by Thomas-Keating Ltd.<sup>9</sup> that provides  $> 50$  dB isolation. Insertion loss is about 6 dB. The isolator is essential to protect the injection source from the FEL power after startup. For this injection-locking system, the FEL is run in a two arm configuration as shown in Fig. 2. A silicon plate, offset slightly from Brewster's angle, couples injection power in and FEL power out. This allows full coupling to the FEL resonator's fundamental mode. The Si plate and the FEL end mirrors are tuned to optimize FEL performance. A second beam-splitter directs about 90% to the users lab. A spectrum analyzer located in the users lab analyzes the FEL output.

The spectrum analyzer is based on a heterodyne detection system. 240 GHz FEL radiation is attenuated

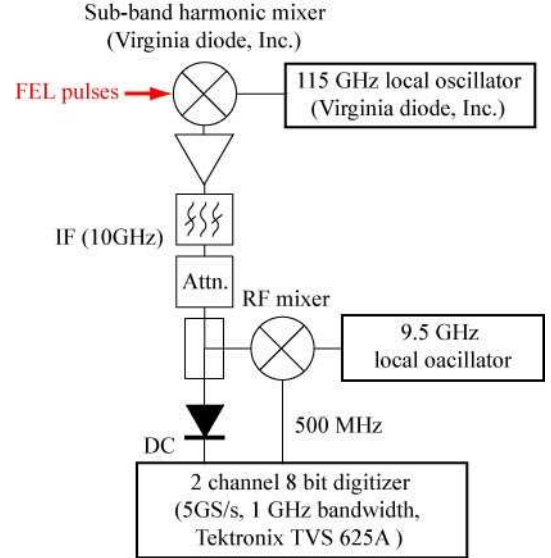


FIG. 3: Circuit diagram of the 240 GHz spectrum analyzer.

and directed to a subharmonically-pumped mixer (VDI) driven at 115 GHz by a second VDI multiplier chain as Local Oscillator. A 10 GHz intermediate frequency (IF) signal is then amplified, filtered, and sent to a second mixer to further downconvert to 500 MHz. A bandpass filter in the IF chain is used to remove negative frequency artifacts. A 2 channel, 8 bit, high performance digitizer (Tektronix TVS 625A) measures both signals and passes the digitized signals to a computer for fast Fourier transform processing. The sampling rate of the digitizer is

5 GS/s and the bandwidth is 1 GHz. The whole spectrum analyzer is controlled by LabView programs which record the single pulse FEL spectra. The IF signal is also detected by a 10 GHz square-law detector.

It is instructive to compare this injection-locking demonstration with a previous experiment in 1991.<sup>10</sup> That experiment used a molecular gas laser which is not tunable as an injection source at 2.5 THz for the UCSB FIR FEL (890 GHz - 4.7 THz). The FEL cavity length had to be adjusted to match the longitudinal mode frequency to the injection source. Evidence for injection-locking was compelling but indirect, as the power spectrum of the emission could not be measured. The tremendous improvements in tunable solid state oscillators in the intervening 16 years have made injection-locked MM-wave FEL at UCSB a robust tool for experimentation.

We are developing a high frequency pulsed EPR spectrometer to investigate the structure and dynamics of biological molecules in aqueous solution. This setup includes the injection-locked FEL, running at 240 GHz, a 9 T superconducting magnet with high homogeneity, a quasi-optical pulse generator<sup>11,12</sup> and quasi-optical delay line<sup>13</sup>. Details of the EPR setup will be described elsewhere.

Currently, most high-power pulsed EPR spectrometers are near the X-band frequency of 9.5 GHz with kW-level power. A trend in the evolution of next generation pulsed EPR is for higher frequency both for a finer spectral resolution and a better resolution for fast structural changes due to less motional averaging.<sup>14</sup> Currently there exist

only a few high-power pulsed EPR spectrometers at frequencies above 50 GHz. A 1 kW Klystron amplifier-based 95 GHz system operates at Cornell.<sup>15</sup> Another Klystron based-pulsed EPR system is being built at St. Andrews UK. A Gyrotron-amplifier may also be useful for high-power pulsed EPR. Although the systems are currently not used for pulsed EPR, 10-15 W 250 GHz and 14 W 140 GHz Gyrotron-based system have been built at MIT.<sup>16,17</sup> Compared with these other sources, the UCSB FEL has significant advantages for pulsed EPR at 240 GHz and higher frequencies. First, the FEL is the most powerful source at 240 GHz. Moreover the UCSB FEL is widely tunable from 120 GHz to 4.7 THz, which covers  $g=2$  EPR signals from 5 T to higher than magnetic fields produced by any existing DC-field magnets.<sup>18</sup> For the UCSB MM-FEL, 240 GHz is at the low end of its frequency range and can produce hundreds of watts of power. It can produce much higher power at higher frequencies. Thus the FEL will work even better for pulsed EPR at a higher frequency.

In summary, we have demonstrated injection-locked FEL operation with sub-MHz bandwidth using a tunable solid state source as an injection seed. The high-power, tunability, and extremely narrow linewidth of the FEL opens up the possibility for new applications such as high-power, high-frequency, pulsed EPR spectroscopy.

We wish to thank David Enyeart for his support of the injection-locking system installation and FEL operation. This work was supported by NSF grants (DMR-0321365 and DMR-0520481).

---

\* Electronic address: [susumu@iqcd.ucsb.edu](mailto:susumu@iqcd.ucsb.edu)

<sup>1</sup> M. S. Sherwin, C. A. Schmuttenmaer, and P. M. Bucksbaum, Report on DOE/NSF/NIH workshop on opportunities in THz science (2004), <http://www.er.doe.gov/bes/reports/abstracts.html#THz>.

<sup>2</sup> P. H. Siegel, IEEE trans. microwave theor. tech. **50**, 910 (2002).

<sup>3</sup> D. You, R. R. Jones, P. H. Bucksbaum, and D. R. Dykaar, Opt. Lett. **18**, 290 (1993).

<sup>4</sup> G. L. Carr, M. C. Martin, W. R. McKinney, K. Jordan, G. R. Neil, and G. P. Williams, Nature **420**, 153 (2002).

<sup>5</sup> R. Köhler, A. Tredicucci, F. Beltram, H. E. Beere, E. H. Linfield, A. G. Davies, D. A. Ritchie, R. C. Iotti, and F. Rossi, Nature **417**, 156 (2002).

<sup>6</sup> 30 mW 240 GHz source and 225-255GHz tunable source were built by Virginia diode Inc. Virginia, USA, <http://www.virginiadiodes.com>.

<sup>7</sup> The University of California Santa Barbara Free-Electron Lasers, <http://sbfel3.ucsb.edu/>.

<sup>8</sup> FELIX - FOM (Rijnhuizen, Netherlands), [http://www.rijnhuizen.nl/research/guthz/felix\\_felice/](http://www.rijnhuizen.nl/research/guthz/felix_felice/).

<sup>9</sup> 240 GHz isolator was built by Thomas Keating Ltd. West Sussex, UK, <http://www.terahertz.co.uk>.

<sup>10</sup> A. Amir, J. F. Knox-Seith, and M. Warden, Phys. Rev. Lett. **66**, 29 (1991).

<sup>11</sup> F. A. Hegmann, J. B. Williams, B. Cole, M. S. Sherwin, J. W. Beeman, and E. E. Haller, Appl. Phys. Lett. **76**, 262 (2000).

<sup>12</sup> M. F. Doty, B. E. Cole, B. T. King, and M. S. Sherwin, Rev. Sci. Instrum. **75**, 2921 (2004).

<sup>13</sup> D. G. Allen et al., to be published.

<sup>14</sup> J. H. Freed, Annu. Rev. Phys. Chem. **51**, 655 (2000).

<sup>15</sup> W. Hofbauer, K. A. Earle, C. R. Dunnam, J. K. Moscicki, and J. H. Freed, Rev. Sci. Instrum. **75**, 1194 (2004).

<sup>16</sup> V. S. Bajaj, C. T. Farrar, M. K. Hornstein, I. Mastovsky, J. Viereg, J. Bryant, B. Eléna, K. E. Kreischer, R. J. Temkin, and R. G. Griffin, J. Magn. Reson. **160**, 85 (2003).

<sup>17</sup> C. D. Joye, R. G. Griffin, M. K. Hornstein, K. N. Hu, K. E. Kreischer, M. Rosay, M. A. Shapiro, J. R. Sirigiri, R. J. Temkin, and P. P. Woskov, IEEE Trans. Plasma Sci. **34**, 518 (2006).

<sup>18</sup> The world's highest DC field magnet is the 45 tesla hybrid magnet in the National High Magnetic Field Laboratory (NHMFL). 45 T corresponds to 1.26 THz for  $g=2$  EPR.